

Translated from Japanese by
SCIENTIFIC TRANSLATION SERVICES
411 Wyntre Lea Dr.
Bryn Mawr, PA 19010

(19) Patent Office of Japan (JP)

(12) Patent Disclosure Bulletin (A)

(11) Patent Disclosure No.: Hei 10-[1998]-101,906

(43) Disclosure Date: April 21, 1998

(51) Int. Cl. ⁶ :	Identification Code:	FI	
C 08 L 63/00		C 08 L 63/00	C
			A
C 08 J 3/20	CFC	C 08 J 3/20	CFC
C 08 K 3/00		C 08 K 3/00	
// (C 08 L 63/00			
C 08 L 83:04)			

Request for Examination: not requested	No. of claims: 2	FD	(Total 11 pages)
--	------------------	----	------------------

(21) Application No.: Hei 8-[1996]-281,748

(22) Application Date: October 3, 1996

(71) Applicant: 000002060
Shinetsu Kagaku Kogyo Co. Ltd.
6-1, 2-chome, Otemach, Chiyoda-ku, Tokyo-to

(72) Inventor: K. Sumida
Silicon Electronic Material Technical Laboratory, Shinetsu Kagaku Kogyo Co. Ltd.
1-10 Hitomi, Oaza, Matsuida-cho, Kakuhyo-gun, Gunma-ken

(72) Inventor: T. Shiohara
Silicon Electronic Material Technical Laboratory, Shinetsu Kagaku Kogyo Co. Ltd.
1-10 Hitomi, Oaza, Matsuida-cho, Kakuhyo-gun, Gunma-ken

(74) Agent: T. Kotori, Patent Attorney (and one other)

(54) [Title of the Invention] METHOD FOR PREPARING A LIQUID EPOXY RESIN COMPOSITION

(57) [Abstract]

[Object] To obtain in an industrially advantageous manner a liquid epoxy resin composition that shows low viscosity and an excellent thin film (gap) penetrating property even when blended with a large amount of inorganic filler and that is suitable as an underfill material for the flip chips of a semiconductor device.

[Solution Means] In preparing a liquid epoxy resin composition in which epoxy resin, curing agent and inorganic filler are its necessary components, the inorganic filler contains 5-30 wt. % of very fine inorganic fillers with specific surface area greater than 4 m²/g and the content of all inorganic fillers accounts for 60 wt. % of the entire composition, a resin composition is formed in the first process in which part of the epoxy resin or part of a mixture of the epoxy resin and the curing agent is added to the inorganic filler that has been prepared to contain more than 50 wt. % of very fine inorganic fillers with specific surface area greater than 4 m²/g and kneaded and the resin

composition thus formed is mixed and kneaded with the remaining filler, epoxy resin and curing agent in the second process.

[Scope of the Patent Claims]

[Claim 1] Method for preparing a liquid epoxy resin composition, characterized in that, in preparing a liquid epoxy resin composition in which epoxy resin, curing agent and inorganic filler are its necessary components, the inorganic filler contains 5-30 wt. % of very fine inorganic fillers with a specific surface area greater than $4 \text{ m}^2/\text{g}$ and the content of all inorganic fillers accounts for 60 wt. % of the entire composition, a resin composition is formed in the first process in which part of the epoxy resin or part of a mixture of the epoxy resin and the curing agent is added to the inorganic filler that has been prepared to contain more than 50 wt. % of very fine inorganic fillers with a specific surface area greater than $4 \text{ m}^2/\text{g}$ and kneaded and the resin composition thus formed is mixed and kneaded with the remaining filler, epoxy resin and curing agent in the second process.

[Claim 2] Method for preparing a liquid epoxy resin composition in accordance with claim 1, in which a silicone modified resin is added in the second process.

[Detailed Description of the Invention]

[0001]

[Field of Industrial Application] The present invention pertains to a method for preparing a liquid epoxy resin composition that shows low viscosity and an excellent gap penetrating property even when blended with a large amount of inorganic filler and that is suitable as an underfill material for the flip chips of a semiconductor device.

[0002]

[Prior Art and Problems to be Solved by the Invention] The conventional liquid epoxy resin composition used as an underfill material especially for the flip chips of a semiconductor contains less than 60 wt. % of inorganic fillers. In recent years, however, in order to make the coefficient of expansion of the liquid epoxy resin composition equal to that of the chips, baseboard and soldering pump, it became necessary to add a large amount of inorganic fillers to the resin composition. The addition of a large amount of inorganic fillers causes an increase in viscosity, a marked decrease in the rate of penetration of the resin composition into the gap between the chip and the baseboard and a marked decrease in the productivity. Therefore, it is desirable to solve such problems.

[0003] In view of the situation mentioned above, the present invention was developed to provide a method for preparing a liquid epoxy resin composition so that a liquid epoxy resin composition with low viscosity and an excellent gap penetrating property even in the presence of organic fillers in a large amount can be prepared with good productivity.

[0004]

[Means for Solving the Problems and Embodiment of Implementation of the Invention] In order to achieve the objects mentioned above, we carried out extensive studies. We found that very fine inorganic fillers and resins could be uniformly and sufficiently wet and a liquid epoxy composition with low viscosity and an excellent thin film penetrating (gap penetrating) property even in the presence of a large amount of inorganic fillers could be obtained when the procedures described below were carried out. In preparing a liquid epoxy resin composition in which epoxy resin, curing agent and inorganic filler are its necessary components, the inorganic filler contains 5-30 wt. % of very fine inorganic fillers with a specific surface area greater than $4 \text{ m}^2/\text{g}$ and the content of all inorganic fillers accounts for 60 wt. % of the entire composition, a resin composition is formed in the first process in which part of the epoxy resin or part of a mixture of the epoxy resin and the curing agent is added to the inorganic filler that has been prepared to contain more than 50 wt. % of very fine inorganic fillers with a specific surface area greater than $4 \text{ m}^2/\text{g}$ and kneaded and the resin composition thus formed is mixed and kneaded with the remaining filler, epoxy resin and curing agent in the second process.

[0005] In order to simultaneously improve the penetration property and reduce expansion of the liquid epoxy resin composition used as the underfill material, it has become a recent trend that very fine fillers with a mean particle

diameter less than 1 micron or preferably less than 0.5 micron are added to the resin composition in a large quantity. Consequently, the specific surface area of the fillers becomes much larger than that of the conventional product, the wetting of the interface between the resin and the fillers becomes extremely poor, the viscosity of the resin composition becomes extremely high and the penetration property becomes low. Such phenomena also take place during the process of kneading the composition. When the amount of fillers blended is greater than 70 wt. %, sometimes the composition becomes hard and preparation of the composition becomes impossible.

[0006] In other words, improvement of the wetting between the resin and the filler surfaces is not possible and the very fine fillers tends to aggregate easily in the low-viscosity liquid epoxy resin composition containing a large amount of very fine fillers with a large specific surface area. Therefore, in the final product, the viscosity of the composition may become very high especially in the region of low shear rate and may not be accompanied by the fluidity, causing the formation of voids and unfilled areas. Moreover, if the aggregates cannot be dispersed, the fillers in the aggregated state may penetrate into the narrow gap, interfering with the fluidity of the fillers, resulting in the formation of non-uniform hardened products and voids. Therefore, the development of techniques for dispersing each individual filler particle in the composition and for uniform wetting of the fine particle surfaces with the resin is very important. We investigated the technique for dispersing the fine particles in the composition and were able to overcome the aforementioned problems by kneading the inorganic fillers with such components as epoxy resin, curing agent, etc., in two processes as mentioned above. This finding enabled us to obtain a liquid epoxy resin composition suitable as an underfill material and led us to develop the present invention.

[0007] Therefore, the present invention can provide a method for preparing a liquid epoxy resin composition with the characteristic procedures described below. In preparing a liquid epoxy resin composition in which epoxy resin, curing agent and inorganic filler are its necessary components, the inorganic filler contains 5-30 wt. % of very fine inorganic fillers with a specific surface area greater than $4 \text{ m}^2/\text{g}$ and the content of all inorganic fillers accounts for 60 wt. % of the entire composition, a resin composition is formed in the first process in which part of the epoxy resin or part of a mixture of the epoxy resin and the curing agent is added to the inorganic filler that has been prepared to contain more than 50 wt. % of very fine inorganic fillers with a specific surface area greater than $4 \text{ m}^2/\text{g}$ and kneaded and the resin composition thus formed is mixed and kneaded with the remaining filler, epoxy resin and curing agent in the second process.

[0008] The present invention is explained in more detail below. The liquid epoxy resin composition according to the present invention is produced by blending epoxy resin, a curing agent, inorganic fillers, etc.

[0009] The epoxy resins that can be used in the present invention can be any well-known epoxy resins as long as they contain more than two epoxy groups in one molecule of resin. Specifically, they include, e.g., bisphenol A type epoxy resins, bisphenol F type epoxy resins, phenol novolak type epoxy resins, cresol novolak type epoxy resins, naphthalene type epoxy resins, biphenyl type epoxy resins, cyclopentadiene type epoxy resins, triphenol alkane type epoxy resins, phenol aralkyl type epoxy resins, etc. Among these epoxy resins, bisphenol A type epoxy resins and bisphenol F type epoxy resins are preferable.

[0010] The total chlorine content of the epoxy resins should be less than 1,500 ppm or preferably less than 1,000 ppm. Moreover, it is desirable that the chlorine content in the water extract obtained when the epoxy resin at a concentration of 50% is extracted with water at 120°C for 20 hours is less than 5 ppm. When the total amount of chlorine is greater than 1,500 ppm and the chlorine content in the water extract is greater than 5 ppm, the moisture resistance reliability of a semiconductor may be lowered in some cases.

[0011] Moreover, powders of the conventional well-known silicone rubbers or gels or heat plasticizable resins made of methyl methacrylate-butadiene-styrene may be added to the composition of the present invention as stress reducing agents.

[0012] The curing agents that can be used in the said epoxy resin compositions include such acid anhydrides as methyl tetrahydrophthalic anhydride, methyl hexahydrophthalic anhydride, hexahydrophthalic anhydride, etc. In addition, such imidazole derivatives as 2-methyl imidazole, 2-ethyl-4-methyl imidazole, 1-cyanoethyl-2-methyl imidazole, 2-phenyl-4,5-dihydroxymethyl imidazole, 2-phenyl-4-methyl-5-hydroxymethyl imidazole, etc., phosphorus type derivatives and cycloamidine type derivatives may be used as the curing agent or the curing promoting agent.

[0013] The amount of the acid anhydride blended as a curing agent should be in the range of 0.3-0.7 mol or preferably 0.4-0.6 mol per one mol of epoxy group in the epoxy resin. When the amount blended is less than 0.3 mol, the hardening property may not be sufficient in some cases while when the amount blended is greater than 0.7 mol, the unreacted acid anhydride will remain, causing a decrease in the glass transition temperature in some cases.

[0014] Furthermore, imidazole derivatives may also be used as curing promoting agents of the acid anhydride type curing agent and also as a curing agent of epoxy resins. When they are used as a curing promoting agent, the amount used should be in the range of 0.01-10 weight parts or preferably 0.5-5 weight parts per 100 weight parts of the total amount of the epoxy resin and the curing agent used. When the amount used is less than 0.01 weight part, the hardening property may be reduced in some cases. When the amount used is greater than 10 weight parts, the hardening property may be excellent but the preservation property may be reduced in some cases.

[0015] In addition, such carboxylic acid hydrazides as dicyanodiamide, adipic acid hydrazide, isophthalic acid hydrazide, etc., may also be used as a curing agent. The amount of carboxylic acid hydrazide used as a curing agent should be in the range of 1-30 weight parts or preferably 5-15 weight parts. When the amount used is less than 1 weight part, the hardening property may not be sufficient in some cases. When the amount used is greater than 30 weight parts, the hardening property may become too high and the preservation property may be reduced in some cases.

[0016] In the present invention, various, conventional, well-known inorganic fillers are added in order to reduce the coefficient of expansion. Molten silica, crystalline silica, boron nitride, aluminum nitride, silicon nitride, magnesia, magnesium silicate, etc., may be used as the inorganic fillers. When the semiconductor element used is an element capable of generating a large quantity of heat, it is desirable to use alumina, boron nitride, aluminum nitride, silicon nitride, etc., with thermal conductivity as large as possible and with small coefficient of expansion are used as fillers. They can be blended with molten silica for use as fillers.

[0017] There is no restriction as to the form of the fillers that can be used. Flake-shaped, dendrite-shaped or sphere-shaped fillers may be used individually or as a mixture. Spherically shaped fillers are especially desirable from the standpoints of low viscosity and fluidity.

[0018] The particle size of the inorganic fillers used depends on the gap between the chip and the baseboard. However, it is desirable that the mean particle size is in the range of 2-30 microns and the maximum particle size is less than 74 microns or preferably less than 50 microns. As for the relationship between the gap and the particle diameter, we found from our experiments that an underfill material with excellent productivity could be obtained when the particle diameter was less than 1/2 of the size of the gap. Therefore, in order to reduce the viscosity of the underfill material and in order to increase the production rate, it is most desirable that the particle size is distributed in the range between the minimum particle size of less than 0.1 micron to the maximum particle size of less than 1/2 of the size of the gap.

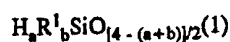
[0019] Furthermore, in the present invention, it is necessary that the inorganic fillers contain 5-30 wt. % or preferably 10-20 wt. % of very fine inorganic fillers with a specific surface area greater than $4 \text{ m}^2/\text{g}$ or mean particle diameter less than 1 micron. When the content is less than 5 wt. %, the viscosity of the composition will increase and the gap penetrating property will decrease rapidly. When the content is greater than 30 wt. %, the viscosity especially under low shear will increase and the gap penetrating property will decrease markedly.

[0020] It is desirable that the amount of the inorganic fillers used is in the range of 150-500 weight parts per 100 weight parts of the total amount of epoxy resin and curing agent used or greater than 60 wt. % or preferably in the range of 60-83 wt. % of the total composition used. When the amount used is less than 60 wt. %, the coefficient of expansion cannot be reduced sufficiently, the water absorption rate will increase and the adhesiveness to the baseboard will become poor. In addition, such problems as detachment between the baseboard or chip and the resin during the temperature cycle, etc., or disruption of the solder pump may occur in some instances. When the amount used is greater than 500 weight parts or greater than 83 wt. %, the viscosity will become too high, the penetration rate of resin will decrease markedly and the productivity will become poor in some cases.

[0021] Moreover, super fine silica particles may be added together with the said inorganic fillers in order to provide the thixotropy to the composition. It is desirable that these fillers have been subject to a surface treatment using a silane coupling agent or a titanium type coupling agent.

[0022] Furthermore, it is desirable that a silicone modified resin, which is a copolymer of organohydrodiene polysiloxane and an alkenyl group containing phenol resin or an alkenyl group containing epoxy resin, is blended into the liquid epoxy resin composition of the present invention, if needed, in order to improve the stress characteristics. In this case, the said silicone modified resins are already well known (Patent Publication No. Sho 63-[1988]-60,069 and Patent Publication No. Sho 63-[1988]-60,070) and copolymers with excellent stress characteristics and with the most suitable structure can be obtained by selecting a proper combination of the structure of the epoxy resin part and the structure of the silicone part. Such well-known copolymers can be used in the present invention.

[0023] Such silicone modified resins include, e.g., copolymers that can be obtained by the addition reaction of alkenyl group containing epoxy resins or alkenyl group containing phenol resins and the SiH group of organohydrodiene polysiloxane that can be represented by the average composition formula (1) given below and that has the number of silicon atoms in the range of 20-400 per molecule and has the number of SiH groups in the range of 1-5.

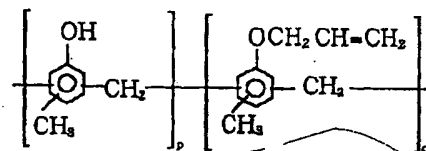
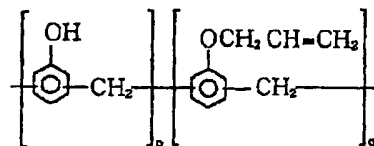
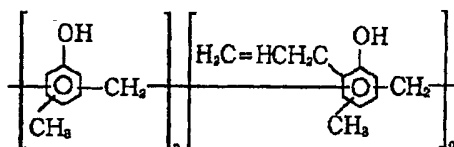
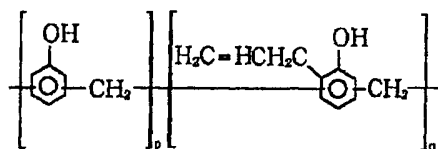
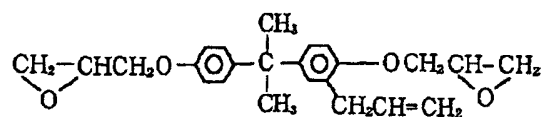
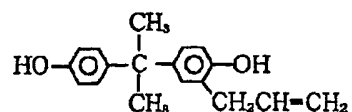
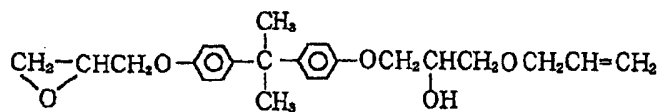
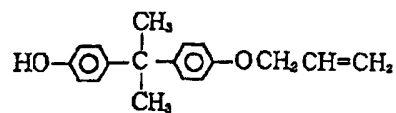


(in which R^1 denotes a substituted or unsubstituted monovalent hydrocarbon group, a and b represent numbers that can satisfy the conditions of $0.02 \leq a \leq 0.1$, $1.8 \leq b \leq 2.2$, $1.8 < a + b \leq 2.3$).

[0024] The alkenyl group containing phenol resins or alkenyl group containing epoxy resins that can be represented by the formula given below are especially suitable for use as the copolymer mentioned above.

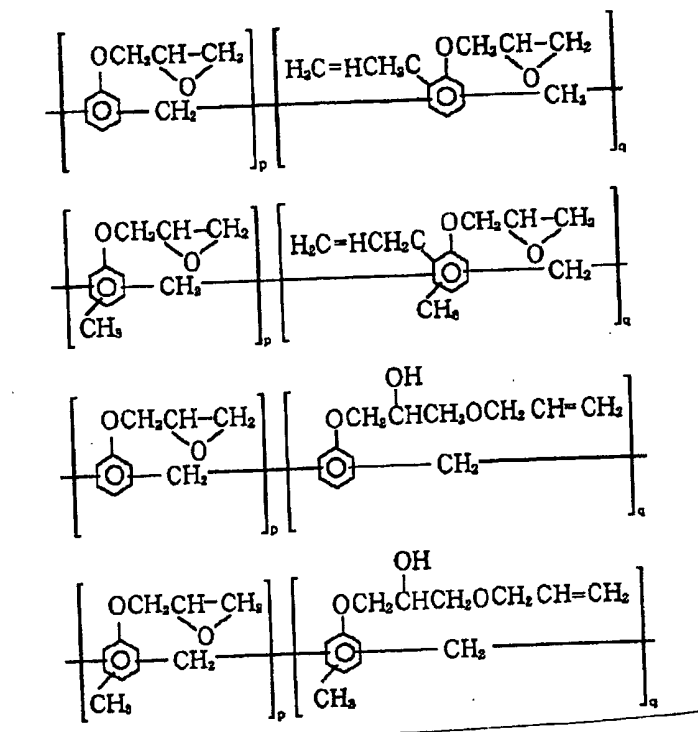
[0025]

[Chemical Formula 1]



[0026]

[Chemical Formula 2]



(p and q usually denote $1 \leq p \leq 20$ or preferably $1 \leq p \leq 10$, $1 \leq q \leq 5$ or preferably $1 \leq q \leq 3$, respectively).

[0027] In the organohydrodiene polysiloxanes of the said formula (1), R^1 usually denotes a substituted or unsubstituted monovalent hydrocarbon group having the number of carbon atoms in the range of 1-12 or preferably having the number of carbons in the range of 1-10 and preferably excluding aliphatic unsaturated bonds. They include, e.g., such alkyl groups as a methyl group, ethyl group, propyl group, butyl group, 2-ethyl butyl group, octyl group, etc., such alkenyl groups as a vinyl group, allyl group, hexenyl group, etc., such cycloalkyl groups as a cyclohexyl group, cyclopentyl group, etc., such aryl groups as a phenyl group, tolyl group, etc., such aralkyl groups as a benzyl group, phenyl ethyl group, etc., the said groups in which part or all of the hydrogen atoms bonded to the carbon atoms are substituted by a halogen atom or a cyano group such as a chloromethyl group, a bromoethyl group, a trifluoropropyl group, etc., a and b represent numbers in the range of $0.002 \leq a \leq 0.1$ or preferably $0.01 \leq a \leq 0.05$ and $1.8 \leq b \leq 2.2$ or preferably $1.8 \leq b \leq 2.0$, respectively, and $1.8 < a+b \leq 2.3$, or preferably $1.81 \leq a+b \leq 2.1$.

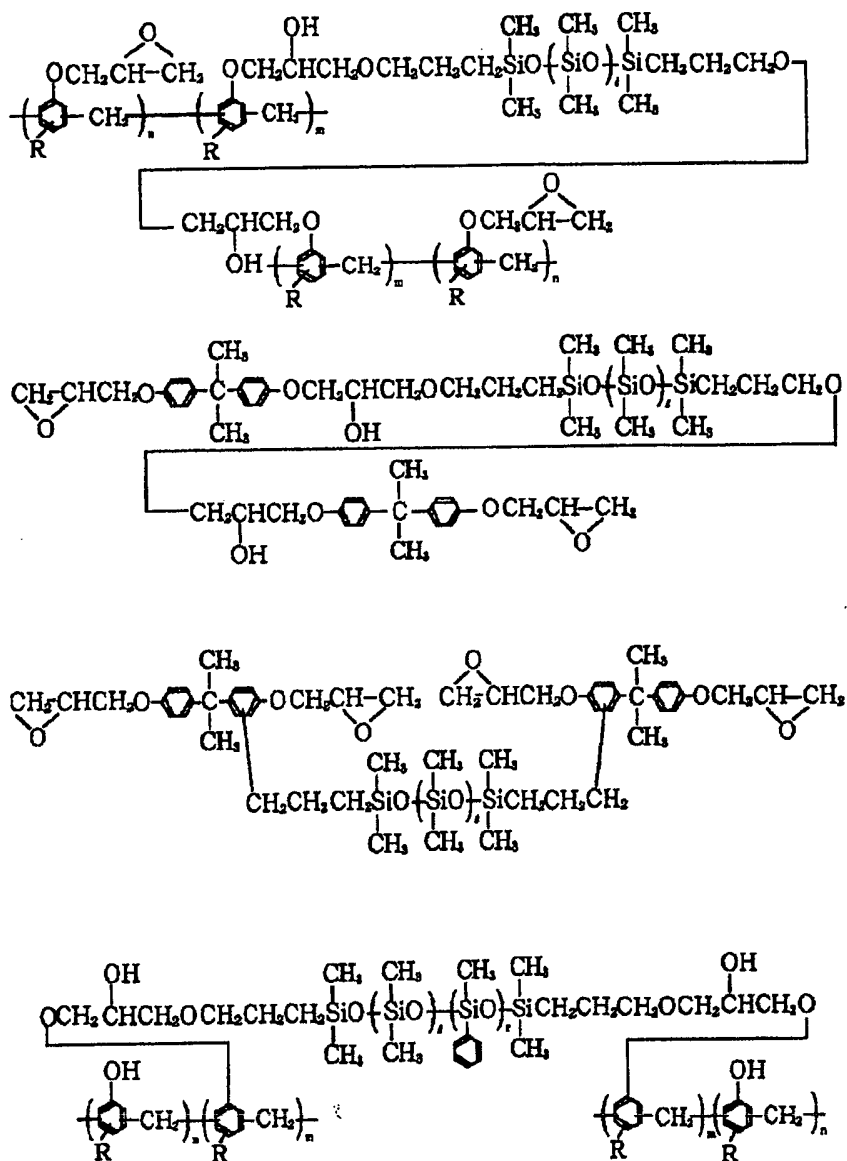
[0028] It is preferable that the number of silicon atoms in one molecule of organohydrodiene polysiloxane is in the range of 20-400 or preferably 40-200 or more preferably 80-150. When the number is less than 20, the silicone modified resin will dissolve in the epoxy resin and the microsea island structure with the epoxy resin as the matrix cannot be formed. Consequently, the low stress property may become absent in some cases. When the number is greater than 400, the silicone modified resin and the epoxy resin may undergo phase separation and the strength of the hardened product may be weakened. In addition, the composition obtained may acquire thixotropy, causing problems when fluidity is required.

[0029] It is desirable that the organohydrodiene polysiloxane used basically has a straight chain structure but it may contain branched chains in part of its structure.

[0030] Representative examples of the silicone modified resins that can be used in the present invention include compounds that can be represented by the structural formula given below.

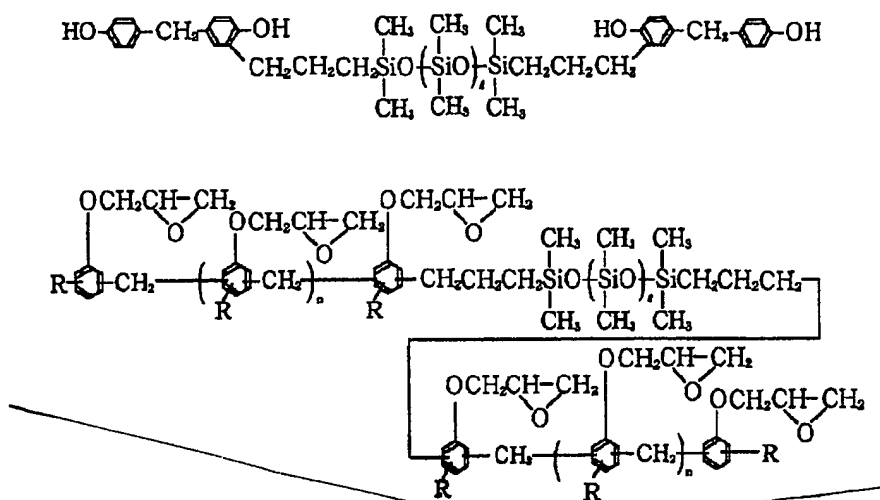
[0031]

[Chemical Formula 3]



[0032]

[Chemical Formula 4]



(in which R denotes a hydrogen atom or a methyl group, l, m, n and r denote numbers in the range of $l = 20-400$ or preferably $40-200$, $m = 0-400$ or preferably $0-200$, $n = 1-20$ or preferably $1-10$ and $r = 1-10$ or preferably $r = 1-3$, respectively).

[0033] The amount of the silicone modified resin blended for 100 weight parts of the sum of the silicone modified resin, epoxy resin and curing agent should be less than 20 weight parts or can be in the range of 0-20 weight parts or preferably 0.5-20 weight parts or more preferably 1-10 weight parts in terms of the amount of the organopolysiloxane contained in the silicone modified resin (e.g., the copolymers mentioned above). When the amount blended is less than 0.5 weight parts, a sufficient low stress property may not be achieved in some cases. When the amount blended is greater than 20 weight parts, the low stress property may be achieved but the viscosity will become very high, the workability will become poor and the strength will decrease in some cases.

[0034] Other optional components, e.g., such silane coupling agents as γ -glycidoxypentyl trimethoxysilane, γ -aminopentyl methoxysilane, γ -glycidoxypentylmethyl dimethoxysilane, γ -aminopentyl triethoxysilane, etc., and such coupling agents as titanium type coupling agents, aluminum type coupling agents, etc., such coloring agents as carbon black, such wetting promoting agents as nonionic type surface active agents, fluorine type surface active agents, silicone oils, etc., antifoaming agents, etc., can be added to the liquid epoxy resin composition of the present invention in some cases. The amounts of such optional components added can be any usual amounts as long as the amounts added will not interfere with the effects of the present invention. The amount of the silane coupling agent added can be in the range of 0.1-3 wt. % or preferably 0.3-1.5 wt. % of the amount of the inorganic filler used. When the amount added is less than 0.1 wt. %, the amount added will not be sufficient to cover the entire surface of the filler and the reinforcing property may not be maintained in some cases. When the amount added is greater than 3 wt. %, the amount added may become excessive and may ooze out onto the surface of the hardened product, producing a poor external appearance in some cases.

[0035] Therefore, in the present invention, in preparing the said liquid resin composition, a resin composition is formed in the first process in which part of the epoxy resin or part of a mixture of the epoxy resin and the curing agent is added to the inorganic filler that has been prepared to contain more than 50 wt. % of very fine inorganic fillers with a specific surface area greater than $4 \text{ m}^2/\text{g}$ and kneaded and the resin composition thus formed is mixed and kneaded with the remaining filler, epoxy resin and curing agent in the second process.

[0036] In this case, the amount of the very fine inorganic fillers with a specific surface area of $4 \text{ m}^2/\text{g}$ used in the first process should be greater than 50 wt. % or preferably is in the range of 70-100 wt. % or more preferably is 100 % of the amount of all inorganic fillers used in the first process. When the amount used is less than 50 wt. %, the surface of the very fine inorganic fillers cannot be sufficiently wetted.

[0037] The amounts of epoxy resins and curing agent added and kneaded into the said inorganic fillers in the first process should be such that the content of the inorganic fillers used in the first process would be greater than 50 wt. % or preferably in the range of 70-80 wt. %. When the content is less than 50 wt. %, shear will not be present and the surface of the fillers cannot be wet sufficiently.

[0038] In the first process, very fine particles alone or inorganic fillers in which very fine particles account for more than 50 wt. % of the fillers are mixed with epoxy resins or a mixture of epoxy resins and a curing agent in such a way that the proportion of the fillers in the mixture will be greater than 50 wt. %. The mixture is kneaded in a kneader capable of simultaneously applying pressure and temperature such as a pressure kneader, a double roll, a triple roll, etc., under pressure and preferably with heating. It is desirable that the kneading temperature is in the range of 10-50°C or preferably 20-30°C. When the temperature is below 10°C, the viscosity cannot be reduced sufficiently and the wetting of the filler surface may not be sufficient in some cases. When the temperature is higher than 50°C, the temperature may become too high and no shear will be present, disrupting aggregation in some cases. The kneading time after the pressure kneading should be in the range of 5-120 minutes or preferably 30-90 minutes.

[0039] In the present invention, the remaining inorganic fillers, epoxy resins, a curing agent, the said silicone modified resins, etc., are weighed and blended into the resin composition that contains the microfine fillers as the main component and that was prepared as described above in the first kneading process. The mixture is mixed and kneaded in a mixing device to obtain the final product. The kneading conditions used in the second process can be identical to those used in the first process.

[0040] Moreover, in the present invention, it is desirable that the said silicone modified resins are added in the second process.

[0041] Furthermore, such conventional, well-known epoxy group containing diluents as *n*-butyl glycidyl ether, phenyl glycidyl ether, styrene oxide, *t*-butyl phenyl glycidyl ether, dicyclopentadiene diepoxide, etc., may be added in the second process in order to lower the viscosity.

[0042] There is no specific limit as to the viscosity of the liquid epoxy resin composition obtained in the present invention. However, it is desirable that the viscosity at 25°C is in the range of 10-5,000 p or preferably 20-3,000 p.

[0043]

[Effect of the Invention] With the use of the method of the present invention for preparing a liquid epoxy resin composition, a liquid epoxy resin composition with low viscosity and excellent thin film (gap) penetrating property even in the presence of a large quantity of blended inorganic fillers can be obtained. The liquid epoxy resin composition thus obtained is suitable for use as the underfill material for the flip chips of a semiconductor device.

[0044]

[Actual Examples] Actual examples and comparison examples will be used to explain the present invention but the present invention is not limited to these actual examples.

[0045] [Actual Examples 1-3 and Comparison Examples 1 and 2] Bisphenol A type epoxy resin (RE410 S, a product of Nippon Kayaku Co.), bisphenol F type epoxy resin (RE303SL, a product of Nippon Kayaku Co.), spherical silica with a mean particle diameter of 0.5 micron and a specific surface area of 7 m²/g (SO25R, a product of Adomatechs Co.), spherical silica with a mean particle diameter of 8 microns and a specific surface area of 1.2 m²/g (SE8, a product of Tokuyama Soda Co.) and a silane coupling agent (γ -glycidioxypropyl trimethoxysilane, KBM 403, a product of Shinetsu Kagaku Kogyo Co. Ltd.) in the proportions given in Table 1 were kneaded in a pressure kneader at a temperature of 25°C and under a pressure of 15 kg/cm² for 60 minutes.

[0046] Next, the master resin composition thus obtained and other components were accurately weighed as shown in Table 2 and were mixed and kneaded in a gate mixer to obtain liquid epoxy resin compositions.

[0047] The liquid epoxy resin compositions thus obtained were evaluated by the methods described below. The results of the evaluation are also shown in Table 2.

Gelation time: The sample was placed on a hot plate heated to 175°C and the time of gelation was measured with the use of a stopwatch.

Viscosity: The viscosity at 25°C was determined with the use of the BH model rotary viscometer at a number of rotations of 4 rpm.

Penetration time: Spencer of about 8 microns was built between two slide glasses measuring 30 x 50 x 1 mm for forming gaps. This test piece was placed on a hot plate at 100°C and the epoxy resin composition was allowed to make thin film penetration into the gaps. The time needed to yield a 20-mm-thick film was measured.

[0048] T_g and coefficient of linear expansion α_1 : A test piece measuring 5 x 5 x 15 mm was used to measure the values at a rate of temperature increase of 5°C per minute with the use of TMA.

[0049]

[Table 1]

Master resin composition (weight parts)		Actual example			Comparison Example	
		1	2	3	1	2
Epoxy resin	RE410S	200		200	200	
	RE303SL		200			200
Inorganic fillers	SO25R	500	500	500	100	
	SE8			100		500
Silane coupling agent	KBM403	5	5	5		

[0050]

[Table 2]

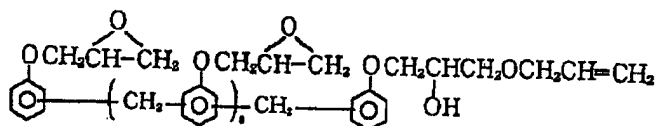
Composition (weight parts)		Actual Example			Comparison Example	
		1	2	3	1	2
Master resin composition		70.5	70.5	80.5	30	70
Epoxy resin	RE303SL		55			55
	RE410S	55		55	55	
Inorganic fillers	SE8	200	200	190	190	150
	SO25R				50	50
Silane coupling agent	KBM403	0.5	0.5	0.5	1.0	1.0
Curing agent*		3	3	3	3	3
Silicone modified resin**		25	25	25	25	25
Carbon black		0.3	0.3	0.3	0.3	0.3
Results of evaluation	Gelation time (sec)	60	60	60	60	60
	Viscosity (p)	1800	1500	2000	3000	2500
	Penetration time (minutes)	3	3	4	10	8
	Coefficient of linear expansion α_1 (ppm/c)	25	25	25	25	25
	Tg (°C)	140	140	140	140	140

*curing agent:

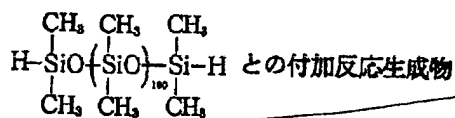
2-phenyl-4,5-dihydroxymethyl imidazole

**silicone modified resin:

Addition product of



and



[0051] The results shown in Table 2 confirm that the rates of thin film (gap) penetration at low viscosity of the liquid epoxy resin compositions (Actual Examples 1-3) obtained by the method of preparation of the present invention are high.